

Form PTO-1390US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE
(Rev. 5-93)

**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371**

ATTORNEY'S DOCKET NO **H 4494 PCT/US**

U S APPLICATION NO (if known sec 17 CFR 1.5)

10/088432

INTERNATIONAL APPLICATION NO.
PCT/EP00/08924

INTERNATIONAL FILING DATE
September 13, 2000

PRIORITY DATE CLAIMED
September 22, 1999

TITLE OF INVENTION

METHOD FOR RESTRUCTURING KERATIN FIBERS

APPLICANT(S) FOR DO/EO/US

Astrid Kleen, Andrea Saettler, Horst Hoeffkes, Ralf Otto, and Oliver Brabaender

Applicant herewith submits to the United States Designated/Elected Office (EO/DO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau)
 - b. ☒ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). **UNEXECUTED**
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern other document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A FIRST preliminary amendment
☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☐ Other items or information:.

Version with Markings to Show Changes Made;
Information Disclosure Citation (Form PTO-1449) and References; and
International Search Report

"Express Mail" mailing label number EL 615775281 US

PATENT
Docket No. H 4494 PCT/US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:
Kleen, et al.

International Application No. PCT/EP00/08924
International Filing Date: September 13, 2000

Serial No. To be assigned **Examiner:** To be assigned
Filed: To be assigned **Art Unit:** To be assigned

Title: METHOD FOR RESTRUCTURING KERATIN FIBERS

"Express Mail Post Office to Addressee" service mailing label number EL 615775281 US

PRELIMINARY AMENDMENT

Box PCT
Assistant Commissioner for Patents
Washington, DC 20231

Attn: DO/EO/US

Sir:

Prior to examining this application, please amend the application as follows:

In the Specification (Using the English Translation):

On page 1 of the English translation, on a separate line between the title and line 1, please insert the following paragraph:

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On a separate page, after page 23, please insert the enclosed Abstract of the Disclosure.

In the Claims

Please cancel Claims 1 to 12, without prejudice.

Please add the following new claims:

- 13. (NEW) A process for restructuring keratin fibers comprising applying to keratin fibers
(a) at least one enzyme having transglutaminase activity; and
(b) at least one active substance having substrate activity for the enzyme.
14. (NEW) The process of claim 13 wherein the enzyme having transglutaminase activity comprises a calcium-independent transglutaminase.
15. (NEW) The process of claim 13 wherein the active substance having substrate activity comprises at least one protein or protein hydrolyzate, or combinations thereof.
16. (NEW) The process of claim 13 wherein the active substance having substrate activity comprises at least one protein or protein hydrolyzate of elastin, collagen, keratin, silk, soya, almond, pea, alga, potato, or wheat, or combinations thereof.
17. (NEW) The process of claim 16 wherein the active substance comprises casein, soya protein or wheat protein, or combinations thereof.
18. (NEW) The process of claim 13 wherein the active substance having substrate activity comprises a substance synthetically functionalized with an H₂N-R group or an H₂N-(CO)-R' group, wherein R and R' represent an unbranched C₁₋₈ alkylene group.

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19. (NEW) The process of claim 18 wherein the synthetically functionalized substance has at least one $\text{H}_2\text{N}-(\text{CH}_2)_4$ group.
20. (NEW) The process of claim 18 wherein the synthetically functionalized substance has at least one $\text{H}_2\text{N}-(\text{CO})-\text{CH}_2-\text{CH}_2$ group.
21. (NEW) The process of claim 13 wherein the enzyme having transglutaminase activity and the active substance having substrate activity are applied simultaneously to the keratin fibers.
22. (NEW) The process of claim 13 wherein the enzyme having transglutaminase activity, and the active substance having substrate activity are applied successively in any order.
23. (NEW) The process of claim 13 wherein the enzyme having transglutaminase activity is contacted with the keratin fibers for a contact time of 3 minutes to 120 minutes.
24. (NEW) A process for setting keratin fibers comprising
 - (a) applying to keratin fibers at least one enzyme having transglutaminase activity and at least one active substance having substrate activity for the enzyme; and
 - (b) setting the keratin fibers.
25. (NEW) The process of claim 24 wherein the active substance having substrate activity comprises at least one protein or protein hydrolyzate of elastin, collagen, keratin, silk, soya, almond, pea, alga, potato, or wheat, or combinations thereof.
26. (NEW) The process of claim 25 wherein the active substance comprises casein, soya protein or wheat protein, or combinations thereof.

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27. (NEW) The process of claim 26 wherein the enzyme having transglutaminase activity comprises a calcium-independent transglutaminase.
28. (NEW) A multi-part kit for restructuring keratin fibers comprising
 - (a) a first composition comprising at least one enzyme having transglutaminase activity; and
 - (b) a second composition having an active substance with substrate activity for the enzyme.
29. (NEW) The kit of claim 28 wherein the active substance having substrate activity comprises at least one protein or protein hydrolyzate of elastin, collagen, keratin, silk, soya, almond, pea, alga, potato, or wheat, or combinations thereof.
30. (NEW) The kit of claim 29 wherein the active substance comprises casein, soya protein or wheat protein, or combinations thereof.
31. (NEW) The kit of claim 30 wherein the enzyme having transglutaminase activity comprises a calcium-independent transglutaminase. --

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REMARKS

Applicants respectfully request the Examiner to enter the above amendments prior to examination of this application.

Status of Claims

Claims 13 to 31 will be pending after entry of the present amendment. Claims 1 to 12 are being canceled without prejudice.

Amendment

The specification is being amended to insert section headers and an abstract of the disclosure in accordance with 37 CFR §1.77 to better conform with US patent practice. The specification is also being amended to insert a cross-reference to related applications in accordance 37 CFR §1.78 and to claim priority to those applications listed therein.

The specification on page 6, line 24 is being amended to delete “coloring” which is a typographical error. No new matter is added as one skilled in the art would recognize that this was an error since the application relates to a method of restructuring hair. Attached hereto is a marked up version of the changes made to the specification entitled “Version With Markings To Show Changes Made.”

New Claims 13 to 31 replace original Claims 1 to 12, and are being presented to better conform with US patent practice. These new claims are supported by the specification for example as shown in the Table below (cites to the specification are for the English translation):

Claim	Support in Specification
13	page 2, lines 6 to 24
14, 27, 31	page 2, lines 26 to 29
15, 16, 17, 25, 26, 29, 30	page 3, lines 20 to 24, page 4, lines 1 to 2
18, 19, 20	page 4, lines 3 to 8
21, 22	page 4, lines 24 to 30
23	page 5, lines 9 to 14
24	page 2, lines 6 to 24
28	page 19, lines 23 to 26

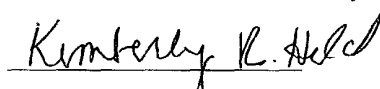
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No new matter is added by the new claims or amendments to the specification.

CONCLUSION

Applicants respectfully request early and favorable notification of allowance of all pending claims. The Assistant Commissioner is authorized to charge any deficiency in the required fee or to credit any overpayment to Deposit Account 01-1250 in connection with this amendment.

Respectfully submitted,



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Abstract of the Disclosure

A method for restructuring keratin fibers is provided. The method includes applying to keratin fibers at least one enzyme having transglutaminase activity and at least one active substance having substrate activity for the enzyme. The enzyme having transglutaminase activity and the active substance having substrate activity for the enzyme can also be applied to keratin fibers for the purpose of setting it.

VERSION WITH MARKINGS TO SHOW CHANGES MADE

In the Specification:

The paragraph on page 6, beginning at line 22 and ending on line 25 has been amended as shown below:

-- However, it has proved to be of advantage in individual cases to select the surfactants from amphoteric or nonionic surfactants because they generally have less influence on the [coloring] process according to the invention. --

On page 22, line 1, the heading "CLAIMS" has been amended as shown below:
[CLAIMS] What is claimed is:

Method for Restructuring Keratin Fibers

This invention relates to a process for restructuring keratin fibers.

Nowadays, human hair is treated in many different ways with hair-care preparations. Such treatments include, for example, the cleaning of hair with shampoos, the care and regeneration of hair with rinses and conditioners and the bleaching, coloring and shaping of hair with coloring and tinting formulations, wave formulations and styling preparations. Among these, formulations for modifying or shading the color of the hair occupy a prominent position. The effect of this behavior is that hair is exposed in various ways to damaging influences which have an adverse effect on the surface structure.

Accordingly, there has been no shortage of attempts to counteract the damage done to the hair structure and to develop restructuring techniques. Various care components have been developed for specific use as aftertreatments for damaged hair. In addition, additional care components have been added to the hair treatment preparations, such as for example the hair fixing lotions used in permanent waving or hair colorants. For example, it was proposed in **DE-A1 196 17 569** that special amino acids be used as care components.

It has surprisingly been found that a distinctly increased restructuring of keratin fibers can be obtained by means of a totally new enzymatic process.

Restructuring in the context of the invention is understood to be a reduction in the damage done to keratin fibers by various influences. For example, restoring the natural strength of the hair plays an important role in this regard. Examples of damaging influences are, for example, permanent wave treatments, oxidative coloring or lightening of the hair and frequent washing, blow drying and combing. Damage can also be done by

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environmental influences, such as UV light for example. Restructured fibers are distinguished, for example, by improved luster, by improved feel and by easier combability. In addition, successful restructuring can be physically manifested as an increase in melting point by comparison with damaged fibers.

In a first embodiment, therefore, the present invention relates to a process for restructuring keratin fibers in which (A) at least one enzyme of the transglutaminase type and (B) at least one active substance with substrate activity for the enzyme are applied to the fibers.

Besides the restructuring of damaged fibers, a second objective of the present invention was to obtain a hair-setting effect for undamaged hair. Accordingly, the process according to the invention may also be used for setting the hair for special styling effects.

Keratin fibers in the context of the invention are understood to include pelts, wool, feathers and in particular human hair.

An enzyme which is preferably used in the process according to the invention is transglutaminase (official name: protein glutamine gamma-glutamyltransferase; EC 2.3.2.13). This enzyme preferentially catalyzes the reaction of the amino acid moiety glutamine in a protein with an alkylamine to form an N5-alkylglutamine protein with release of ammonia. A preferred natural alkylamine which plays a role in this reaction is the amino acid lysine or the amino acid moiety lysine in a protein.

In principle, any enzymes with transglutaminase activity are suitable for carrying out the present invention. Suitable enzymes of this type are, for example, transglutaminases obtained from guinea pig liver, *Physarum polycephalum*, *Medicago sativa* or *Bacillus subtilis*. The calcium-independent transglutaminases described, for example, in **EP 726 317 A2** and in **EP 397 606 A1** which are marketed by Ajinomoto are particularly preferred. The Ajinomoto products Activa® WM and EB are preferred, Activa® WM being particularly preferred.

The use of transglutaminases in cosmetic formulations is already known from the literature. For example, **US 5,490,980** describes a composition for treating human skin, hair or nails with which active substances containing a primary amino group are added onto the glutamine components of the skin, hair or nails by transglutaminase. However, there is nothing in this document which points to the subject of the present invention or to the restructuring properties of this process.

In the context of the invention, an active substance with substrate activity is any substance which can be added onto the hair by the transglutaminase. This can be done, for example, by crosslinking the active substances with substrate activity with one another, i.e. by forming a kind of membrane around the hair. However, this can also be done with advantage by covalent bonding of the active substances with substrate activity to the lysine and/or glutamine components of the hair.

In a first preferred embodiment of the present invention, naturally occurring substances are used as active substances with substrate activity. Proteins, protein hydrolyzates and derivatives thereof are particularly suitable for this purpose. Protein hydrolyzates are product mixtures obtained by acid-, base- or enzyme-catalyzed degradation of proteins.

According to the invention, proteins and protein hydrolyzates of both vegetable and animal origin may be used.

Animal proteins are, for example, elastin, collagen, keratin, silk and milk protein. Examples of proteins of vegetable origin are soya, almond, pea, alga, potato and wheat protein.

Although proteins as such are preferably used, other natural active substances with substrate activity, such as for example peptides, amino acids and corresponding derivatives, may also be used instead. Derivatives of the protein hydrolyzates, for example in the form of their fatty acid condensation products or cationic derivatives, may also be used but are less preferred.

Casein, soya protein and wheat protein are particularly preferred, casein being most particularly preferred.

In a second preferred embodiment of the invention, substances synthetically functionalized with an H_2N-R group or an $H_2N-(CO)-R'$ group, where R and R' stand for an unbranched C_{1-8} alkylene group, are used as active substances with substrate activity. Particularly preferred functional groups are the groups $H_2N-(CH_2)_4$ and $H_2N-(CO)-CH_2-CH_2-$ derived from lysine or glutamine.

In addition, monomers such as, for example, lysine and glutamine may be used in accordance with the invention as active substances with substrate activity. They may be used both as an additional active substance with substrate activity and as sole component. In a preferred embodiment of the invention, both proteins and corresponding monomers may be used during the process to improve fastness to washing by fairly rapidly building up a dense network of the active substances with substrate activity.

The active substances with substrate activity are present in the preparations used in accordance with the invention in quantities of preferably 0.005 to 10% by weight, based on the composition as a whole. Quantities of 0.01 to 2% by weight are particularly preferred. The ratio by weight of the transglutaminase type enzyme to the active substance with substrate activity is preferably 1:4000 to 1:1 and more preferably 1:2000 to 1:50.

So far as the time sequence of the process is concerned, the invention is not subject to any limitations. It is possible in principle to apply two separate preparations containing (a) the active substance with substrate activity and (b) the transglutaminase type enzyme successively in any order to the fibers. In a preferred embodiment, the two components (a) and (b) are applied to the fibers in that order. According to the invention, the components may also be separately applied in the order (b) \rightarrow (a). In

this case, however, the time interval between steps (a) and (b) should not be too long to ensure that the fibers do not dry between the steps.

Although this two-stage process does produce the desired effects, it may be preferred to carry out the process according to the invention in a single stage because single-stage processes are easier to carry out. The active substance with substrate activity may be applied together with the enzyme preparation. In a preferred embodiment of the invention, the two components are mixed immediately before application.

Although in principle the preparation can remain on the hair, a preferred embodiment of the process according to the invention is characterized in that the preparation containing the enzyme is rinsed out after a contact time of 3 to 120 minutes. The preparation may be rinsed out with clean water. Contact times of 15 to 30 minutes have proved to be sufficient in most cases.

Irrespective of the course of the process according to the invention, it has proved to be of advantage to apply the enzyme preparation at a temperature of 20 to 55°C and more particularly at a temperature of 35 to 50°C.

In principle, there are no limits to the nature of the enzyme preparation. According to the invention, aqueous, alcoholic and oily preparations and mixtures thereof are particularly suitable. Aqueous preparations are particularly preferred. These may be, for example, solutions, dispersions, emulsions (water-in-oil emulsions, oil-in-water emulsions and multiple emulsions and PIT emulsions). The pH value of these preparations is generally in the range from 2 to 10, preferably in the range from 4 to 9 and more preferably in the range from 6 to 8.

In a preferred embodiment of the present invention, the enzyme preparations are formulated as a thickened solution. To this end, the preparations are thickened with thickeners, such as agar agar, guar gum, alginates, xanthan gum, gum arabic, karaya gum, locust bean gum, linseed

gums, dextrans, cellulose derivatives, for example methyl cellulose, hydroxyalkyl cellulose and carboxymethyl cellulose, starch fractions and derivatives, such as amylose, amylopectin and dextrans, clays, for example bentonite, or fully synthetic hydrocolloids, for example polyvinyl alcohol or even polyacrylic acid polymers. In a preferred embodiment, the enzyme preparations are formulated with low viscosities.

Besides the enzyme and optionally the active substance with substrate activity, the enzyme preparations may contain all the usual constituents suitable for the treatment of keratin fibers, particularly human hair. Aqueous preparations are preferred. Aqueous preparations in the context of the invention are preparations which contain at least 50% by weight water, based on the preparation as a whole.

It has proved to be of advantage for the enzyme preparation to contain at least one surfactant. Suitable surfactants are both anionic, ampholytic, zwitterionic or nonionic surfactants and cationic surfactants. If necessary, the expert may carry out simple preliminary tests to determine whether the various surfactants have any effect on the activity of the transglutaminase type enzyme.

A preferred embodiment of the invention is characterized by the use of a combination of anionic and nonionic surfactants or a combination of anionic and amphoteric surfactants.

However, it has proved to be of advantage in individual cases to select the surfactants from amphoteric or nonionic surfactants because they generally have less influence on the coloring process according to the invention.

Suitable anionic surfactants in the preparations according to the invention are any anionic surface-active substances suitable for use on the human body. Such substances are characterized by a water-solubilizing anionic group such as, for example, a carboxylate, sulfate, sulfonate or phosphate group and a lipophilic alkyl group containing around 10 to 22

carbon atoms. In addition, glycol or polyglycol ether groups, ester, ether and amide and hydroxyl groups may also be present in the molecule.

Nonionic surfactants contain, for example, a polyol group, a polyalkylene glycol ether group or a combination of polyol and polyglycol ether groups as the hydrophilic group. Examples of such compounds are

- products of the addition of 2 to 30 moles of ethylene oxide and/or 0 to 5 moles of propylene oxide onto linear fatty alcohols containing 8 to 22 carbon atoms, onto fatty acids containing 12 to 22 carbon atoms and onto alkylphenols containing 8 to 15 carbon atoms in the alkyl group,
- C₁₂₋₂₂ fatty acid monoesters and diesters of products of the addition of 1 to 30 moles of ethylene oxide to glycerol,
- C₈₋₂₂ alkyl mono- and oligoglycosides and ethoxylated analogs thereof and
- products of the addition of 5 to 60 moles of ethylene oxide to castor oil and hydrogenated castor oil.

Preferred nonionic surfactants are alkyl polyglycosides corresponding to the general formula R¹O-(Z)_x. These compounds are characterized by the following parameters.

- The alkyl group R¹ contains 6 to 22 carbon atoms and may be both linear and branched. Primary linear and 2-methyl-branched aliphatic groups are preferred. Such alkyl groups are, for example, 1-octyl, 1-decyl, 1-lauryl, 1-myristyl, 1-cetyl and 1-stearyl. 1-Octyl, 1-decyl, 1-lauryl and 1-myristyl are particularly preferred. Where so-called "oxo alcohols" are used as starting materials, compounds with an odd number of carbon atoms in the alkyl chain predominate.

- The alkyl polyglycosides suitable for use in accordance with the invention may, for example, contain only one particular alkyl group R¹. However, such compounds are normally prepared from natural fats and oils or mineral oils. In this case, mixtures corresponding to the starting

compounds or corresponding to the particular working up of these compounds are present as the alkyl groups R.

Particularly preferred alkyl polyglycosides are those in which R¹ consists

- 5 - essentially of C₈ and C₁₀ alkyl groups,
- essentially of C₁₂ and C₁₄ alkyl groups,
- essentially of C₈ to C₁₆ alkyl groups or
- essentially of C₁₂ to C₁₆ alkyl groups.

Any mono- or oligosaccharides may be used as the sugar unit Z.

- 10 Sugars containing 5 or 6 carbon atoms and the corresponding oligosaccharides are normally used. Examples of such sugars are glucose, fructose, galactose, arabinose, ribose, xylose, lyxose, allose, altrose, mannose, gulose, idose, talose and sucrose. Preferred sugar units are glucose, fructose, galactose, arabinose and sucrose; glucose is particularly
- 15 preferred.

The alkyl polyglycosides suitable for use in accordance with the invention contain on average 1.1 to 5 sugar units. Alkyl polyglycosides with x values of 1.1 to 1.6 are preferred. Alkyl glycosides where x is 1.1 to 1.4 are most particularly preferred.

- 20 Besides acting as surfactants, the alkyl glycosides may also be used to improve the fixing of perfume components to the hair. Accordingly, in cases where the effect of the perfume oil on the hair is intended to last longer than the duration of the hair treatment, alkyl glycosides will preferably be used as another ingredient of the preparations according to
- 25 the invention.

Alkoxyated homologs of the alkyl polyglycosides mentioned may also be used in accordance with the invention. These homologs may contain on average up to 10 ethylene oxide and/or propylene oxide units per alkyl glycoside unit.

Zwitterionic surfactants may also be used, particularly as co-surfactants. In the context of the invention, zwitterionic surfactants are surface-active compounds which contain at least one quaternary ammonium group and at least one $\text{-COO}^{(-)}$ or $\text{-SO}_3^{(-)}$ group in the molecule.

5 Particularly suitable zwitterionic surfactants are the so-called betaines, such as N-alkyl-N,N-dimethyl ammonium glycinate, for example cocoalkyl dimethyl ammonium glycinate, N-acylaminopropyl-N,N-dimethyl ammonium glycinate, for example cocoacylaminopropyl dimethyl ammonium gly-
 10 8 to 18 carbon atoms in the alkyl or acyl group and cocoacylaminoethyl hydroxyethyl carboxymethyl glycinate. A preferred zwitterionic surfactant is the fatty acid amide derivative known by the CTFA name of Cocamidopropyl Betaine.

Also suitable, particularly as co-surfactants, are ampholytic
 15 surfactants. Ampholytic surfactants are surface-active compounds which, in addition to a C_{8-18} alkyl or acyl group, contain at least one free amino group and at least one -COOH or $\text{-SO}_3\text{H}$ group in the molecule and which are capable of forming inner salts. Examples of suitable ampholytic
 20 surfactants are N-alkyl glycines, N-alkyl propionic acids, N-alkyl aminobutyric acids, N-alkyl iminodipropionic acids, N-hydroxyethyl-N-alkyl amidopropyl glycines, N-alkyl taurines, N-alkyl sarcosines, 2-alkyl aminopropionic acids and alkyl aminoacetic acids containing around 8 to 18 carbon atoms in the alkyl group. Particularly preferred ampholytic
 25 surfactants are N-cocoalkyl aminopropionate, cocoacyl aminoethyl aminopropionate and C_{12-18} acyl sarcosine.

According to the invention, the cationic surfactants used are particularly those of the quaternary ammonium compound, esterquat and amidoamine type.

Preferred quaternary ammonium compounds are ammonium
 30 halides, more particularly chlorides and bromides, such as alkyl trimethyl

The surfactants representing addition products of ethylene and/or
30 propylene oxide with fatty alcohols or derivatives of these addition products

may be both products with a "normal" homolog distribution and products with a narrow homolog distribution. Products with a "normal" homolog distribution are mixtures of homologs which are obtained in the reaction of fatty alcohol and alkylene oxide using alkali metals, alkali metal hydroxides or alkali metal alcoholates as catalysts. By contrast, narrow homolog distributions are obtained when, for example, hydrotalcites, alkaline earth metal salts of ether carboxylic acids, alkaline earth metal oxides, hydroxides or alcoholates are used as catalysts. The use of products with a narrow homolog distribution can be of advantage.

10 In addition, the enzyme preparations used in accordance with the invention preferably contain at least one oil component.

Oil components suitable for the purposes of the invention are, in principle, any water-insoluble oils and fatty compounds and mixtures thereof with solid paraffins and waxes. According to the invention, water-insoluble substances are defined as substances of which less than 0.1% by weight dissolves in water at 20°C. The melting point of the individual oil or fatty components is preferably below about 40°C. Oil and fatty components which are liquid at room temperature, i.e. below 25°C, can be particularly preferred for the purposes of the invention. However, where several oil and fatty components and optionally solid paraffins and waxes are used, it is generally sufficient if the mixture of the oil and fatty components and optionally paraffins and waxes satisfies these requirements.

25 A preferred group of oil components are vegetable oils. Examples of such oils are sunflower oil, olive oil, soya oil, rapeseed oil, almond oil, jojoba oil, orange oil, wheatgerm oil, peach kernel oil and the liquid fractions of coconut oil.

However, other triglyceride oils, such as the liquid fractions of bovine tallow, and synthetic triglyceride oils are also suitable.

30 Another group of compounds particularly preferred for use as oil

components in accordance with the invention are liquid paraffin oils and synthetic hydrocarbons and di-n-alkyl ethers containing a total of 12 to 36 carbon atoms and, more particularly, 12 to 24 carbon atoms, such as for example di-n-octyl ether, di-n-decyl ether, di-n-nonyl ether, di-n-undecyl ether, di-n-dodecyl ether, n-hexyl-n-octyl ether, n-octyl-n-decyl ether, n-decyl-n-undecyl ether, n-undecyl-n-dodecyl ether and n-hexyl-n-undecyl ether and ditert.butyl ether, diisopentyl ether, di-3-ethyldecyl ether, tert.butyl-n-octyl ether, isopentyl-n-octyl ether and 2-methylpentyl-n-octyl ether. The compounds 1,3-di-(2-ethylhexyl)-cyclohexane and di-n-octyl ether obtainable as commercial products (Cetiol® S and Cetiol® OE, respectively) can be preferred.

Other oil components suitable for use in accordance with the invention are fatty acid and fatty alcohol esters. The monoesters of fatty acids with alcohols containing 3 to 24 carbon atoms are preferred. This group of substances are products of the esterification of fatty acids containing 8 to 24 carbon atoms such as, for example, caproic acid, caprylic acid, 2-ethylhexanoic acid, capric acid, lauric acid, isotridecanoic acid, myristic acid, palmitic acid, palmitoleic acid, stearic acid, isostearic acid, oleic acid, elaidic acid, petroselic acid, linoleic acid, linolenic acid, elaeostearic acid, arachic acid, gadoleic acid, behenic acid and erucic acid and the technical mixtures thereof obtained, for example, in the pressure hydrolysis of natural fats and oils, in the reduction of aldehydes from Roelen's oxosynthesis or in the dimerization of unsaturated fatty acids with alcohols such as, for example, isopropyl alcohol, caproic alcohol, caprylic alcohol, 2-ethylhexyl alcohol, capric alcohol, lauryl alcohol, isotridecyl alcohol, myristyl alcohol, cetyl alcohol, palmitoleyl alcohol, stearyl alcohol, isostearyl alcohol, oleyl alcohol, elaidyl alcohol, petroseliny alcohol, linolyl alcohol, linolenyl alcohol, elaeostearyl alcohol, arachyl alcohol, gadoleyl alcohol, behenyl alcohol, erucyl alcohol and brassidyl alcohol and the technical mixtures thereof obtained, for example, in the high-pressure

hydrogenation of technical methyl esters based on fats and oils or aldehydes from Roelen's oxosynthesis and as monomer fraction in the dimerization of unsaturated fatty alcohols. According to the invention, iso-propyl myristate, isononanoic acid C₁₆₋₁₈ alkyl ester (Cetiol® SN), stearic
5 acid-2-ethylhexyl ester (Cetiol® 868), cetyl oleate, glycerol tricaprylate, cocofatty alcohol caprate/caprylate and n-butyl stearate are particularly preferred.

Other oil components suitable for use in accordance with the invention are dicarboxylic acid esters, such as di-n-butyl adipate, di-(2-ethylhexyl)-adipate, di-(2-ethylhexyl)-succinate and diisotridecyl acelate,
10 and diol esters, such as ethylene glycol dioleate, ethylene glycol diisotridecanoate, propylene glycol di-(2-ethylhexanoate), propylene diisostearate, propylene glycol dipelargonate, butanediol diisostearate and neopentyl glycol dicaprylate, and also complex esters, for example diacetyl glycerol
15 monostearate.

Finally, fatty alcohols containing 8 to 22 carbon atoms may also be used as oil components in accordance with the invention. The fatty alcohols may be saturated or unsaturated and linear or branched. Examples of fatty alcohols suitable for use in accordance with the invention
20 are decanol, octanol, octenol, dodecenol, decenol, octadienol, dodecadienol, decadienol, oleyl alcohol, erucyl alcohol, ricinoyl alcohol, stearyl alcohol, isostearyl alcohol, cetyl alcohol, lauryl alcohol, myristyl alcohol, arachidyl alcohol, capryl alcohol, capric alcohol, linoleyl alcohol, linolenyl alcohol and behenyl alcohol and Guerbet alcohols thereof (this list
25 is purely exemplary and is not intended to limit the invention in any way). However, the fatty alcohols emanate from preferably natural fatty acids, normally being obtained from the esters of the fatty acids by reduction. According to the invention, it is also possible to use the fatty alcohol cuts which are produced by reduction of naturally occurring triglycerides, such
30 as bovine tallow, palm oil, peanut oil, rapeseed oil, cottonseed oil, soybean

– cationic guar derivatives such as, in particular, the products marketed under the names of Cosmedia® Guar and Jaguar®,

According to the invention, preferred cationic polymers are quaternized cellulose derivatives, polymeric dimethyl diallyl ammonium salts, Polyquaternium 27 and copolymers thereof and polymers of the
30 Polyquaternium 2 type. Cationic cellulose derivatives, more particularly the

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- quaternized amines, such as methyl-1-alkylamidoethyl-2-alkylimidazolium methosulfate,
- defoamers, such as silicones,
- dyes for coloring the composition,
- 5 - antidandruff agents, such as Piroctone Olamine, Zinc Omadine and Climbazol,
- sun protection factors, more particularly derivatized benzophenones, cinnamic acid derivatives and triazines,
- substances for adjusting the pH value, such as for example the usual
- 10 acids, more particularly edible acids and bases,
- active principles, such as allantoin, pyrrolidone carboxylic acids and salts thereof and bisabolol,
- vitamins, provitamins and vitamin precursors, more particularly those of groups A, B₃, B₅, B₆, C, E, F and H,
- 15 - plant extracts, such as the extracts of green tea, oak bark, stinging nettle, hamamelis, hops, camomile, burdock root, horse willow, hawthorn, lime blossom, almond, aloe vera, pine needle, horse chestnut, sandalwood, juniper, coconut, mango, apricot, lemon, wheat, kiwi, melon, orange, grapefruit, sage, rosemary, birch, mallow, lady's
- 20 smock, creeping thyme, yarrow, thyme, balm, restharrow, coltsfoot, hibiscus, meristem, ginseng and ginger root ,
- cholesterol,
- consistency factors, such as sugar esters, polyol esters or polyol alkyl ethers,
- 25 - fats and waxes, such as spermaceti, beeswax, montan wax and paraffins,
- fatty acid alkanolamides,
- complexing agents, such as EDTA, NTA, β -alanine diacetic acid and phosphonic acids,
- 30 - swelling and penetration agents, such as glycerol, propylene glycol

monoethyl ether, carbonates, hydrogen carbonates, guanidines, ureas and primary, secondary and tertiary phosphates,

- opacifiers, such as latex, styrene/PVP and styrene/acrylamide copolymers,
- 5 - pearlizers, such as ethylene glycol mono- and distearate and PEG-3-distearate,
- pigments,
- stabilizers for hydrogen peroxide and other oxidizing agents,
- propellents, such as propane/butane mixtures, N₂O, dimethyl ether,
- 10 CO₂ and air.

Information on other optional components and the quantities in which they are used can be found in the reference books known to the expert, for example Kh. Schrader, **Grundlagen und Rezepturen der Kosmetika, 2nd Edition, Hüthig Buch Verlag, Heidelberg, 1989.**

- 15 In a second embodiment, the present invention relates to the use of (A) at least one enzyme of the transglutaminase type and (B) at least one active substance with substrate activity for the enzyme for restructuring keratin fibers.

- 20 In a third embodiment, the present invention relates to the use of (A) at least one enzyme of the transglutaminase type and (B) at least one active substance with substrate activity for the enzyme for setting keratin fibers.

- 25 In a fourth embodiment, the present invention relates to a two-part kit for restructuring keratin fibers which contains a first preparation containing (a) an active substance with substrate activity and a second composition containing (b) an enzyme of the transglutaminase type.

The following Examples are intended to illustrate the invention.

Examples

a) Pretreatment

Alkinco tresses (0.5 g, Code 6634) were subjected to a conventional permanent wave treatment with the commercial product "Poly Lock-Normale Dauerwelle". In the first step of this permanent wave treatment, the fibers were exposed to the reducing solution (containing 7.9% by weight thioglycolic acid) for 40 minutes at room temperature, rinsed with clean water and then fixed for 10 minutes at room temperature (oxidizing solution containing 2.6% by weight of hydrogen peroxide). After the oxidative treatment, the fibers were rinsed and dried.

b) Aftertreatment

Test 1): aftertreatment according to the invention

15 The tresses were immersed for 60 minutes at a temperature of 50°C in 2 ml of an aqueous casein solution (30 mg/ml, adjusted to pH 7.6 with tris(hydroxymethyl)aminomethane (TRIS) HCl buffer) to which 100 µl of an aqueous transglutaminase solution (50 mg/ml Activa® WM¹, corresponding to 0.5 mg/ml active substance, adjusted to pH 7.6 with TRIS HCl buffer) had been added.

20 1 powder form commercial product, 1% by weight transglutaminase in
99% by weight dextrin

Test 2): aftertreatment with casein

25 The tresses were immersed for 60 minutes at a temperature of 50°C
in 2 ml of an aqueous casein solution (30 mg/ml, adjusted to pH 7.6 with
tris(hydroxymethyl)aminomethane (TRIS) HCl buffer)

Test 3): aftertreatment with transglutaminase

The tresses were immersed for 60 minutes at a temperature of 50°C
30 in 2 ml of an aqueous transglutaminase solution (1.2 mg/ml Activa® WM,

corresponding to 0.012 mg/ml active substance, adjusted to pH 7.6 with TRIS HCl buffer).

c) Determination of the hair-structuring effect by HP-DSC

The following melting points were determined by DSC analysis (Perkin Elmer DSC-7):

	Mean value in °C	Scatter
Test 1)	148.24	0.40
Test 2)	147.91	0.07
Test 3)	147.77	0.18
Test 4)	147.62	0.02
Tresses with no aftertreatment		

Statistical evaluation by a two-sided heterogeneous T-Test revealed a significance of 99.73% between the measured values of tests 1) and 2).

CLAIMS

1. A process for restructuring keratin fibers, characterized in that
 - (A) at least one enzyme of the transglutaminase type and
 - (B) at least one active substance with substrate activity for the enzymeare applied to the fibers.
2. A process as claimed in claim 1, characterized in that the enzyme is
5 a calcium-independent transglutaminase.
3. A process as claimed in claim 1 or 2, characterized in that the active substance with substrate activity is a protein or a protein hydrolyzate.
4. A process as claimed in claim 3, characterized in that the active substance with substrate activity is selected from casein, soya protein and
10 wheat protein.
5. A process as claimed in claim 1 or 2, characterized in that the active substance with substrate activity is an active substance synthetically functionalized with an $\text{H}_2\text{N-R}$ group or an $\text{H}_2\text{N}(\text{CO})\text{-R}'$ group, where R and R' stand for an unbranched C_{1-8} alkylene group.
- 15 6. A process as claimed in claim 5, characterized in that the active substance with substrate activity carries at least one $\text{H}_2\text{N}(\text{CH}_2)_4$ group.
7. A process as claimed in claim 5, characterized in that the active substance with substrate activity carries at least one $\text{H}_2\text{N}(\text{CO})\text{-CH}_2\text{-CH}_2$ group.
- 20 8. A process as claimed in any of claims 1 to 7, characterized in that the enzyme preparation is applied to the fibers together with the active substance with substrate activity.
9. A process as claimed in any of claims 1 to 8, characterized in that the contact time is 3 to 120 minutes.
- 25 10. The use of (A) at least one enzyme of the transglutaminase type and (B) at least one active substance with substrate activity for the enzyme for restructuring keratin fibers.
11. The use of (A) at least one enzyme of the transglutaminase type and

12. A two-part kit for restructuring keratin fibers, characterized in that it contains (a) a composition containing an active substance with substrate activity and (b) a composition containing an enzyme of the transglutaminase type.

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(12) NACH DEM VERTRAG ÜBER DIE INTERNATIONALE ZUSAMMENARBEIT AUF DEM GEBIET DES
PATENTWESENS (PCT) VERÖFFENTLICHTE INTERNATIONALE ANMELDUNG

(19) Weltorganisation für geistiges Eigentum
Internationales Büro



(43) Internationales Veröffentlichungsdatum
29. März 2001 (29.03.2001)

PCT

(10) Internationale Veröffentlichungsnummer
WO 01/21139 A3

(51) Internationale Patentklassifikation⁷: **A61K 7/06**

(21) Internationales Aktenzeichen: **PCT/EP00/08924**

(22) Internationales Anmeldedatum:
13. September 2000 (13.09.2000)

(25) Einreichungssprache: **Deutsch**

(26) Veröffentlichungssprache: **Deutsch**

(30) Angaben zur Priorität:
199 45 487.6 22. September 1999 (22.09.1999) **DE**

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(84) Bestimmungsstaaten (regional): europäisches Patent (AT,
BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC,
NL, PT, SE).

Veröffentlicht:

— mit internationalem Recherchenbericht

(88) Veröffentlichungsdatum des internationalen
Recherchenberichts: **16. August 2001**

Zur Erklärung der Zweibuchstaben-Codes, und der anderen
Abkürzungen wird auf die Erklärungen ("Guidance Notes on
Codes and Abbreviations") am Anfang jeder regulären Ausgabe
der PCT-Gazette verwiesen.

(54) Title: **METHOD FOR RESTRUCTURING KERATIN FIBERS USING AN ENZYME OF THE TRANSGLUTAMINASE
TYPE**

(54) Bezeichnung: **VERFAHREN ZUR RESTRUKTURIERUNG KERATINISCHER FASERN ENZYM VOM TYP DER
TRANSGLUTAMINASE EINSETZEND**

(57) Abstract: The invention relates to a method for restructuring keratin fibers. According to the inventive method, (A) at least one enzyme of the transglutaminase type and (B) at least one active substance that has a substrate activity for said enzyme are applied on the fibers. The inventive method allows the restructuring of damaged fibers.

(57) Zusammenfassung: Die vorliegende Erfindung betrifft ein Verfahren zur Restrukturierung keratinischer Fasern, bei dem auf die Fasern (A) mindestens ein Enzym vom Typ der Transglutaminase und (B) mindestens ein Wirkstoff, der eine Substrataktivität für das Enzym aufweist, aufgebracht werden. Mithilfe dieses Verfahrens können geschädigte Fasern restrukturiert werden.

WO 01/21139 A3

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PTO/SB/01 (6-95)

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0010/PTO Rev. 6/95 <div style="text-align: center;">DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION</div> <div style="display: flex; justify-content: space-around; margin-top: 10px;"> <input type="checkbox"/> Declaration Submitted with Initial Filing OR <input checked="" type="checkbox"/> Declaration Submitted after Initial Filing </div>	<div style="text-align: center;">U.S. Department of Commerce Patent and Trademark Office</div> <table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 50%;">Attorney Docket Number</td> <td style="width: 50%;">H 4494 PCT/US</td> </tr> <tr> <td>First Named Inventor</td> <td>Kleen, Astrid</td> </tr> <tr> <td colspan="2" style="text-align: center;"><i>COMPLETE IF KNOWN</i></td> </tr> <tr> <td>Application Number</td> <td>10/088,432</td> </tr> <tr> <td>Filing Date</td> <td></td> </tr> <tr> <td>Group Art Unit</td> <td></td> </tr> <tr> <td>Examiner Name</td> <td></td> </tr> </table>	Attorney Docket Number	H 4494 PCT/US	First Named Inventor	Kleen, Astrid	<i>COMPLETE IF KNOWN</i>		Application Number	10/088,432	Filing Date		Group Art Unit		Examiner Name	
Attorney Docket Number	H 4494 PCT/US														
First Named Inventor	Kleen, Astrid														
<i>COMPLETE IF KNOWN</i>															
Application Number	10/088,432														
Filing Date															
Group Art Unit															
Examiner Name															

As a below named inventor, I hereby declare that

My residence, post office address, and citizenship are as stated below next to my name

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

METHOD FOR RESTRUCTURING KERATIN FIBERS

(Title of the Invention)

the specification of which

☐ is attached hereto

OR

☒ was filed on (MM/DD/YYYY) 09/13/2000 as United States Application Number or PCT International

Application Number PCT/EP00/08924 and was amended on (MM/DD/YYYY) _____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT International application having a filing date before that of the application on which priority is claimed

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached?	
				YES	NO
199 45 487.6	DE	09/22/1999	<input type="checkbox"/>	<input type="checkbox"/>	<input checked="" type="checkbox"/>
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Application Number(s)	Filing Date (MM/DD/YYYY)	
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DECLARATION

Page 2

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s), or §365© of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112 1 acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)
	PCT/EP00/08924	09/13/2000	

☐ Additional U.S. or PCT international application numbers are listed on a supplemental priority sheet attached hereto.

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.

☐ Firm Name Customer Number or label
OR

☒ List Attorney(s) and/or agent(s) name and registration number below:

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor:

☐ A petition has been filed for this unsigned inventor

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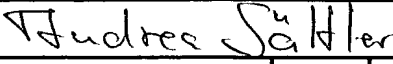
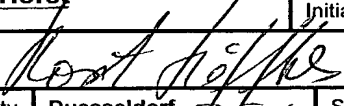

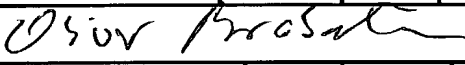
H 4494 PCT/US

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DECLARATION					ADDITIONAL INVENTOR(S) Supplemental Sheet				
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<input type="checkbox"/> Additional inventors are being named on supplemental sheet(s) attached hereto									